Efficiency Enhancement of Cu(In,Ga)Se₂ Solar Cells by Applying SiO₂–PEG/PVP Antireflection Coatings

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An effective approach was presented to enhance photoelectric conversion efficiency of Cu(In,Ga)Se₂ (CIGS) solar cells by using modified SiO₂ antireflection coatings (ARCs) to harvest more incident sunlight. Polyethylene glycol (PEG) and polyvinyl pyrrolidone (PVP) used as additives were introduced into silica sols to prepare SiO₂–PEG and SiO₂–PVP coatings in the sol–gel dip-coating process, respectively. The different effects of PEG and PVP additives on SiO₂ coatings were analyzed and the antireflection performance of SiO₂–PEG and SiO₂–PVP coatings was investigated. The transmittance over 97% ranging from 450 nm to 700 nm with a maximum transmittance over 99.40% at about 550 nm was achieved for both SiO₂–PEG2000A and SiO₂–PVP0.5 coatings. The relative efficiencies of CIGS solar cells coated with SiO₂–PEG2000A and SiO₂–PVP0.5 ARCs were increased by 7.27% and 8.33%, respectively. The modified SiO₂ ARCs possessed the advantages of the low manufacturing cost, good adhesion, superior antireflective performance and the feasible method for large area fabrication.

KEY WORDS: CIGS solar cells; Efficiency; Antireflection coatings; Sol–gel process; Polymeric additive

1. Introduction

Solar cells have attracted much attention as a potential renewable energy source in the past decade and are expected to grow even faster in the future^[1–5]. Among various solar cells,

thin-film solar cells, such as Cu(In,Ga)Se₂ (CIGS), CdTe, etc, possess significant potential as the next generation of photovoltaic devices^[6–10]. Especially, CIGS solar cells have been regarded as one of the most promising thin film solar cells due to their highest efficiency, low manufacturing costs and long-term reliability^[7–13].

Many efforts have been devoted to developing photovoltaic devices that are nano-architected to achieve higher device efficiency and lowering the manufacturing costs by the use of traditional processing technique. The solar incidence of light suffers from the Fresnel reflection on the surface of window layer of CIGS solar cells^[14–17], resulting in the deficiency of the photogenerated current. Therefore, the reduction in reflection of incident light is generally essential to increase the efficiency of solar cells. A conventional way widely utilized to suppress reflectance is depositing single or multilayer films onto the window layer of solar cells to form antireflection coatings (ARCs)^[18–20]. However, the performance of the ARCs is depending on the refractive index of substrates and angles of incidence of light, and the antireflection band is limited in a narrow range. By the theoretical and experimental investigation, the array architecture with sub-wavelength size is considered to enhance broadband and omnidirectional effect of ARCs. And a great variety of fabrication methods, such as electron beam lithography and dry etching, have been used to form a refractive-index-graded structure^[11, 21–25], however, the high cost and complication of fabrication process and limit of large-area fabrication are their serious drawbacks. Consequently, ARCs with cost-effectiveness, broadband and easy fit for large-area fabrication may still retain a challenge in solar cell applications.

The preparation of SiO₂-polymer composite coatings by one step sol–gel technique is also an effective method to achieve a refractive-index-graded structure, which has been focused on modification of coatings and applications in high power lasers^[26–30]. In this paper, an effective approach to increase the photovoltaic efficiency of CIGS solar cells by using modified SiO₂ ARCs to harvest more incident sunlight was reported. Polyethylene glycol (PEG) and polyvinyl pyrrolidone (PVP) used as additives were introduced into the silica sols to prepare SiO₂-PEG and SiO₂-PVP coatings, respectively. The different effects of PEG and PVP additives on the structure and mechanical properties of the SiO₂ coatings were analyzed. The antireflection performances of SiO₂-PEG and SiO₂-PVP coatings coated CIGS solar cells were investigated. The excellent antireflection performance of the modified SiO₂ ARCs and its advantage of preparation process might make for a more streamlined process for practical application of CIGS solar cells.

2. Experimental

All chemical reagents in the experiment were purchased from Sinopharm Chemical Reagent Co. Ltd. and used without further purification. CIGS solar cells with structure of glass/Mo/CIGS/CdS/i-ZnO/ZnO:Al/Ag grids were fabricated with active area of 0.4 cm² as shown in [Scheme 1](#), and described in details in Ref.^[31]. CdS fabricated by chemical bath deposition and i-ZnO and ZnO:Al layer deposited by RF magnetron sputtering served as buffer layers and window layers, respectively. The CIGS solar cells were simply cleaned before fabrication of coatings with ethanol followed by N₂ drying process.

The silica precursor sol was synthesized using tetraethyl orthosilicate (TEOS) as precursor, ammonia as catalyst and ethanol as solvent with a molar ratio of 1:2:40. The stable and transparent silica sol was obtained via removing the catalyst by refluxing process at 80 °C after aging for 7 days. To prepare SiO₂-PEG and SiO₂-PVP precursor sols, the corresponding additives of PEG ($M_w=2000\&10000$) and PVP ($M_w=1300000$) were used. PEG or PVP was dissolved in ethanol, and then TEOS and ammonia were added to the ethanol solution of PEG or PVP under stirring condition for 2 h. The molar ratio of PEG and TEOS is 1:1 and the contents of PVP are 0.1, 0.3, 0.5, 1.0 and 1.5 wt%.

The SiO₂-PEG and SiO₂-PVP coatings were prepared by sol-gel dip-coating technique (Dip Coater, WPTL0.01, MTI Co., China). The cleaned substrates were dipped into the sols and kept for 2 min, and then drew from the sols with a controllable withdrawal rate (controlled speed range from 3 to 200 mm/min) to obtain the desired thickness, followed by drying in oven at 50 °C for 15 min. The substrates used in the experiments were glass slides and CIGS solar cells. The prepared coatings from SiO₂ sols containing PEG with molecule weight of 2000 and 10000 were named as SiO₂-PEG2000 and SiO₂-PEG10000, respectively. After these coating were annealed in oven at 400 °C for 30 min, they were named as SiO₂-PEG2000A and SiO₂-PEG10000A. The obtained coatings from SiO₂ sols containing PVP with contents of 0.1, 0.3, 0.5, 1.0 and 1.5, were marked as SiO₂-PVP0.1, SiO₂-PVP0.3, SiO₂-PVP0.5, SiO₂-PVP1.0 and SiO₂-PVP1.5, respectively.

The morphology of the coatings was characterized by field emission high resolution scanning electron microscopy (FE-SEM, JSM-6700F). The surface structure of sols was examined by transmission electron microscopy (TEM, JEM2100F). The refractive indices and extinction coefficients of the coatings prepared on silicon substrates were measured by means of spectroscopic ellipsometer (SC620UVN). A spectrophotometer (Hitachi U3010) was used to record transmittance spectra of coatings from the visible to the near infrared region at room

temperature equipped with an integrating sphere using BaSO₄ as the reference, and an appendix to measure the reflectance spectra of coatings, where the samples were measured at normal incidence. The photovoltaic (PV) performance parameters of CIGS solar cells were measured under a simulated illumination (Solar simulator: Newport Oriel 93194A) with a light intensity of 100 mW/cm².

3. Results and Discussion

3.1. Morphology and structure

The surface structure of silica sols was investigated by TEM and the results are shown in Fig. 1(a), which shows highly branched particular microstructure with size under 20 nm. A large amount of amorphous SiO₂ particles are formed due to the faster speed of condensation than that of hydrolysis and network with cross-link microstructure of SiO₂ particles is obtained during the sol-gel process^[32]. The formation of SiO₂ coatings by dip-coating technique is a process of gelation by connection of the particles, and the FE-SEM images of SiO₂ coatings are shown in Fig. 1(b). A porous microstructure consisting of agglomerated nanoparticles with the average particle size of about 20 nm and nanopores are observed. Most of the pores in coatings have a size less than 50 nm but some of them with a distributed range from 100 to 200 nm, indicating the rugged porous structure.

Fig. 2(a) shows the surface morphology of SiO₂-PEG2000 coatings. During hydrolysis and condensation process of TEOS, SiO₂ particles with many residual hydroxyls on the surface are gradually produced. And the dissolved PEG chains possess hydroxyls and form hydrogen bonds with hydrolyzed TEOS, which modifies the surface properties of SiO₂ particles and also improves the condensation process of TEOS. During dip-coating process, the interaction between the special chemical group of PEG and SiO₂ particles leads to modification of the microstructure of SiO₂ coatings. After annealing of the samples at 400 °C, most of the covered PEG was removed, which lead to the formation of a porous structure as shown in Fig. 2(b). By comparing with that of SiO₂ coatings, it can be seen that the additive of PEG is effective to create relatively uniform porous structure. The test results of mechanical performance by using tape to removing the coating from substrate indicate that the modified porous coatings possess better adhesion properties than that of unmodified one, which arises from increasing contact area between SiO₂ particles and substrate. When the molecule weight of PEG increases, the coating with high porosity (shown in Fig. 2(c)) arising from generated larger pores is obtained.

The influence of additive of PVP has been investigated. Fig. 3 shows the surface morphology

of SiO₂–PVP0.5 coatings. The network microstructure consisting of modified SiO₂ particles with smaller size than that in SiO₂ coatings can be seen. The interaction of group of pyrrolidone in side-chains of PVP molecule and surface hydroxyls of TEOS form the hydrogen bonds, which considerably modifies the properties of silica sols during the sol–gel process. The SiO₂ particles can be completely covered with PVP by forming linkage on the surface, and therefore are restricted by PVP to form dispersed nanoparticles. The condensation of PVP covered SiO₂ particles are weakened due to the existence of shell-like PVP layer, which results in formation of the smaller nanoparticles than that in pure silica sols. During the dip-coating process, the PVP covered SiO₂ particles were firstly deposited on the substrates to form a denser layer and a porosity-graded structure can be achieved from bottom to top of coatings. The denser layer can improve the adhesion due to increasing contact area and interaction between PVP molecular and substrate.

The appropriate refractive index and the optical film thickness are very important for optical coatings to obtain high antireflection performance. The refractive index depends mainly on the ratio of silica to void, while other factors, such as dip-coating speed, viscosity of sol and speed of evaporation of solvents, affect the coating thickness. The equation^[33], $\text{Porosity} = 1 - (n^2 - 1)/(n_d^2 - 1)$, can give the correlation between refractive index and porosity of coatings, where n and n_d are the refractive indices of the resultant coating and dense material, respectively. The modification of silica sol by introducing PEG or PVP during sol–gel process can influence the microstructure as discussed above and therefore adjust the porosity of the coatings. From Fig. 4(a), it can be seen that the refractive index of the SiO₂–PEG2000 coating shows a bigger value than that of SiO₂ coating due to the further condensation of PEG covered SiO₂ particles to form larger and denser particles. During annealing process, the decomposition of PEG occurred and a lot of pores were created resulting in high porosity structure and controllable refractive index. By adding PEG additive with higher molecular weight ($M_w=10000$), the resultant coating has higher porosity and lower refractive index. The refractive indices of PVP modified coatings are shown in Fig. 4(b) versus the additive content of PVP. With increasing content of PVP, the refractive index of coating increases, indicating the denser structure and lower porosity of the coatings.

3.2. Optical property

To determine the antireflection performance, the transmittance and reflectance spectra of SiO₂–PEG coatings on glass substrates were measured according to the method in Ref.^[34], and

the results are shown in Fig. 5. It can be seen from Fig. 5(a) that the SiO₂-PEG2000A coatings show a high transmittance over the whole solar spectrum with a maximum value of 99.40% at about 550 nm responding to the highest intensity of distribution of solar energy. The average solar transmittance of SiO₂-PEG2000A, SiO₂-PEG10000A and SiO₂-PEG2000 coatings and uncoated substrates are 94.84%, 93.60%, 90.71% and 89.44% in the wavelength range from 400 to 2500 nm, respectively. Fig. 5(b) shows the corresponding reflectance spectra of the coatings versus the incident wavelength over the solar spectrum. The reflectance of the SiO₂-PEG2000A coatings is clearly reduced and the minimum value in visible region is 0.23%. Such obviously reduced reflectance of the substrates and the improvement of the adhesion have shown their advantage for practical applications.

Antireflection performance of SiO₂-PVP coatings over the solar spectrum was investigated, by comparing with uncoated substrate and SiO₂ single coating, and the transmittance and reflectance spectra of the coatings are shown in Fig. 6. In Fig. 6(a), there is a region with a transmittance higher than 98% for SiO₂-PVP0.5 coatings ranged from 450 to 700 nm, and the maximum transmittance of 99.44% appears at about 550 nm. The average of solar transmittance of SiO₂-PVP0.5, and SiO₂ single coatings in wavelength range from 400 to 2500 nm are 96.15%, and 94.08%, respectively. The higher transmittance of the SiO₂-PVP0.5 coatings over the whole solar spectrum and broaden the band testify that the addition of PVP in SiO₂ coatings is more advantageous to achieve the high antireflection performance. The transmittance of coatings with different content of PVP was investigated and the maximum values as a function of the content of PVP are shown in Fig. 6(b), in which the parabola-like change appears and achieves the optimal value at the content of 0.5 wt%. Fig. 6(c, d) shows the corresponding reflectance spectra of coatings as a function of the incident wavelength over the solar spectrum. The reflectance of the substrates is obviously reduced by using coatings with different content of PVP additive and achieves a better effect than that of SiO₂ coatings.

3.3. Photovoltaic performance

To demonstrate the effect of ARCs on solar energy power conversion efficiency (PCE), CIGS solar cells coated with and without ARCs were prepared. The AZO used in CIGS solar cells as window layer have a transparency of about 88% in the visible range, indicating more than 10% incident light reflected, which is similar to that of glass substrates. The preparation of ARCs on AZO is carried out to increase the transmittance and the efficiency of CIGS solar cells by harvesting more incident light. The current-voltage (I - V) characteristics of the

ARC-coated CIGS devices comparing with the uncoated CIGS samples were investigated. The results of efficiency enhancement of CIGS solar cells by using SiO₂-PEG/PVP ARCs can be seen from Fig. 7(a), where four different CIGS solar cells were used as substrates and measured for each coating. The average values of measured photovoltaic parameters are listed in Table 1 (see supporting information) and the PCE of cells are all increased comparing with that of uncoated cells by introducing ARCs. The I - V characteristic of SiO₂-PEG2000A coated CIGS solar cells is shown in Fig. 7(b), and a short-circuit photocurrent density (J_{sc}) of 35.69 mA/cm² with an open-circuit voltage (V_{oc}) of 508.7 mV and a calculated fill factor (FF) of 60.91% have been obtained, respectively. The overall PCE (η) of CIGS is increased from 10.31% to 11.06%. Fig. 7(b) also shows the result of SiO₂-PVP0.5 coated CIGS solar cells and the overall η of CIGS is increased from 10.20% to 11.05%. It can be seen that generally J_{sc} is increased by introducing ARCs. In other words, the improvement of the CIGS solar cells is mainly attributed to the increase of J_{sc} , because more photonics coupled into the adsorbing layer in CIGS solar cells increase carrier concentration and improve photogenerate current. These results indicate that the ARCs are more advantageous to enhance the photovoltaic performance of CIGS solar cells due to more solar incidence with suitable energy coupled into the cells.

The external quantum efficiency (EQE) is the ratio between the current obtained outside the solar cells and the number of incoming photons, which depends on both the absorption of light and the collection of charges. The EQE of CIGS solar cells coated with SiO₂-PEG2000A and SiO₂-PVP0.5 were investigated by comparing with the responding uncoated CIGS solar cells, and the results are shown in Fig. 7(c). A flat spectral response in wavelength range from about 510 to 990 nm can be observed and increased by 6%–8% by using SiO₂-PEG2000A coatings. For SiO₂-PVP0.5 coated CIGS solar cells, the increase with a value of 7%–10% in the same wavelength range is achieved. The results discussed here indicate that more light absorption can improve the photovoltaic performance of devices. The introduction of ARCs onto CIGS solar cells provides a promising way to increase the efficiency of the devices. Modifying ARCs by adding both PEG and PVP are effective method to adjust the structure of the coatings and enhance its mechanical and antireflection performance.

4. Conclusion

In summary, photoelectric conversion of CIGS solar cells have been enhanced by using SiO₂-PEG and SiO₂-PVP ARCs with a modified microstructure and improved mechanical

adhesion via one stage synthetic method. PEG was introduced into silica sols during sol–gel process to create relatively uniform porous structure and adjustable void ration. PVP was dispersed throughout the silica sols system to form the refractive-index-graded structure during fabrication of the coatings. Both PEG and PVP can enhance adhesion between the coatings and the substrates. Very high transmittance over 97% ranging from 450 to 700 nm with a maximum transmittance over 99.40% at about 550 nm was achieved for both SiO₂–PEG2000A and SiO₂–PVP0.5 coatings. The relative efficiencies of CIGS solar cells after coating with SiO₂–PEG2000A and SiO₂–PVP0.5 ARCs are increased by 7.27% and 8.33%, respectively. The investigation results testified that the SiO₂–PEG/PVP ARCs were suitable alternative ARCs with the excellent antireflection performance for solar cells devices. In addition, the preparation procedure of the modified SiO₂ ARCs is simple, cost-effective and more conducive to large-area fabrication and practical application in solar cells.

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Figure and table captions

Scheme 1 Schematic of CIGS solar cell on soda-lime glass with ARCs.

Fig. 1 (a) TEM image of SiO₂ sol, (b) SEM image of SiO₂ coating.

Fig. 2 SEM images of coatings of SiO₂–PEG2000 (a), SiO₂–PEG2000A (b) and SiO₂–PEG10000A (c).

Fig. 3 SEM image of SiO₂–PVP0.5 coatings. The inset is the magnification with a bar scale of 20 nm.

Fig. 4 (a) Refractive index of coatings of: (i) SiO₂, (ii) SiO₂–PEG2000, (iii) SiO₂–PEG2000A and (iv) SiO₂–PEG10000A; (b) plot of refractive index and SiO₂–PVP coatings with different contents of PVP.

Fig. 5 Transmittance (a) and reflectance (b) spectra of coatings of SiO₂–PEG2000, SiO₂–PEG2000A and SiO₂–PEG10000A, comparing with uncoated substrates.

Fig. 6 Transmittance (a) and reflectance (c) spectra of SiO₂–PVP coatings with different content of PVP and uncoated substrates. The maximum transmittance (b) and minimum reflectance (d) of SiO₂–PVP coatings with different content of PVP.

Fig. 7 (a) Efficiency enhancement of CIGS solar cells by applying SiO₂–PEG/PVP ARCs. (b) *I*–*V* characteristic and (c) external quantum efficiency of SiO₂–PEG2000A and SiO₂–PVP0.5 coated CIGS solar cells, comparing with responding uncoated CIGS solar cells.

Figure list:

Scheme 1

Fig. 1

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Fig. 7